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EFFECT OF NONEQUILIBRIUM PHONONS ON  
SUPERCONDUCTING STATES WITH TWO COEXISTING ENERGY GAPS

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ABSTRACT

It is shown that under the influence of tunnel currents, superconductors may exhibit a "first order transition" to a state with two coexisting energy gaps. An energy and number conserving approximation for the collision operator is used to explicitly take into account in the theory the effect of nonequilibrium phonons. Quantitative predictions for experiments are presented.

1. INTRODUCTION

We have now reached, mainly through the study of radiation stimulated superconductivity and of inhomogeneous states, the stage where many "nonequilibrium phases" (nonequilibrium collective effects) may appear in superconductors, immediately bringing to the forefront the problems of stability of dissipative states which are nowadays under intense investigation<sup>1</sup> in the field of nonequilibrium statistical mechanics in general. Thanks to the work of Eckern, Schmid, Schmutz and Schön<sup>2</sup> (ESSS) we have now a large number of cases where the stability of dissipative states has been investigated, including some examples where detailed balance is violated.

Although the work of ESSS predicts many phenomena which can be qualitatively checked with experiment, a quantitative comparison is difficult, mainly because phonons were assumed to remain in equilibrium, a condition which we know is violated in most experimental situations.<sup>3</sup>

In this paper, we return to one of the first examples (which is now part of the more general picture of ESSS) where the ideas of Schmid<sup>4</sup> on the stability of nonequilibrium superconducting states were applied. In ref. 5 (hereafter I) it was shown that in superconducting tunnel junctions, two values of the energy gap could simultaneously be a solution of the gap equation. The results are in qualitative accord with the pioneering experimental work of Dynes, Narayanamurti and Gurno<sup>6</sup> and of Gray and Willemsen<sup>7</sup>.

This paper reports on how the theory of I is modified when nonequilibrium phonons are included in the simplest possible way. This allows us to set limits on the domain of validity of the theory of I and to make quantitative predictions in the regime where we can show that nonequilibrium phonons only renormalize the temperature entering the Landau-Ginzburg equation. If these simple theoretical predictions can be checked quantitatively by experiments, it will give strong support to the theoretically sound global

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picture emerging from the work of ESSS. However, if a quantitative comparison with experiment is not successful, the theoretical calculation of at least the effect of nonequilibrium phonons will need to be improved.

## 2. MICROSCOPIC THEORY

For convenience, we first summarize the results already obtained in refs. 5 and 2. We consider a tunnel junction consisting of the superconductor of interest (the probe) coupled to a second superconductor (the injector). We momentarily neglect the effect of non-equilibrium phonons. We also assume that,

- a) the probe is thin enough that we may assume a uniform injection over the thickness of the junction;
- b) the gap of the injector  $\Delta_i$ , in contrast to the gap of the probe, is not appreciably perturbed by the tunneling process. This can be achieved by using an injector which is much thicker than the probe. Qualitatively, our results should also apply to a more symmetrical case<sup>7</sup> but since it is hard experimentally to make the injector and the probe identical, we prefer to study the case where they are markedly different;
- c) we restrict ourselves to temperatures  $T$  close to the transition temperature  $T_c$  (Landau-Ginzburg region), ( $\Delta, \Delta_i \ll T$ ).

The equations describing such a system have been derived many times. The gap equation is,

$$(\alpha - \beta \Delta^2) \Delta = -\chi \Delta - \xi^2 \nabla^2 \Delta \quad (1)$$

where  $\xi^2$  is the usual Landau Ginzburg coherence length,  $\alpha = (T_c - T)/T_c$ ,  $\beta = 7\zeta(3)/8\pi^2 T^2$  and the "gap control" (or anomalous term, or control function)  $\chi$  is defined by,

$$\chi = - \int_{-\infty}^{\infty} dE \frac{1}{E} N_1(E) \delta n(E) \quad (2)$$

where  $\delta n(E)$  is the angular average (isotropic part) of the energy dependent deviation of the distribution function from its local equilibrium value.  $N_1(E) \equiv \Theta(|E| - \Delta) |E| / (E^2 - \Delta^2)^{1/2}$  is the BCS density of states and  $\delta n(E)$  obeys a Boltzmann equation.<sup>2</sup> The collision operator  $I_{ep}$  can be written down in the relaxation time approximation,  $I_{ep}[\delta n(E)] = \tau_E^{-1} \delta n(E)$  where  $\tau_E$  can be taken as a constant  $\tau_0$  equal to its value at  $E = 0$  and  $T = T_c$  because  $E$  and  $\Delta$  are small with respect to  $T$ . To order  $(\Delta/T)^2$  the scattering-in term can be neglected. Finally, the term representing the effect of tunnel injection can be derived using, for example, golden rule arguments. Since only the part of  $\delta n(E)$  which is odd in energy contributes to  $\chi$  in Eq. (2), we can write,

$$\langle \dot{n}_E \rangle_D = 2B \left\{ N_1^1(E - eV) [n_T(E - eV) - n_T(E)] + (eV \leftrightarrow -eV) \right\} \quad (3)$$

$$B \equiv (8e^2 R \Omega N(0))^{-1}$$

where  $R$  is the resistance of the junction,  $\Omega$  the (effective) volume of the probe,  $N(0)$  the normal density of states and  $N_1^1$  the injector BCS density of states.

It was shown by ESSS that the most stable states in this situation were homogeneous so that the analysis of I applies under more general conditions. The global stability of the various homogeneous stationary states can be determined from the following potential (generalized free energy) which can be obtained from the stationary solutions of the Fokker-Planck equation <sup>2,4</sup> obeyed by our system,

$$F = -2N(0) \int d^3r \int_0^\Delta d\Delta' [\alpha - \beta\Delta'^2 + \chi(\Delta')] \Delta'. \quad (4)$$

Since such a potential function (generalized free energy) exists, our system obeys the principle of detailed balance<sup>1</sup> and its behavior will be analogous to systems in thermodynamic equilibrium; in particular it will exhibit a first order phase transition when two minima of  $F$  correspond to the same value of the generalized free energy density<sup>8</sup>.

To proceed any further, we need an explicit expression for  $\chi$ , the gap control. The Boltzmann equation for the distribution function in the stationary homogeneous situation reduces to  $\delta n(E) =$

$\tau_0(n_E) D$ . The quantity  $\chi$  is computed using this equation and Eqs. (2), (3). The first order expansion in  $\frac{eV}{T}$  of Eq. (3) is plotted in Fig. 1. This graph of  $\delta n^{(1)}(E)$  is useful to understand the results given in analytical form in I.

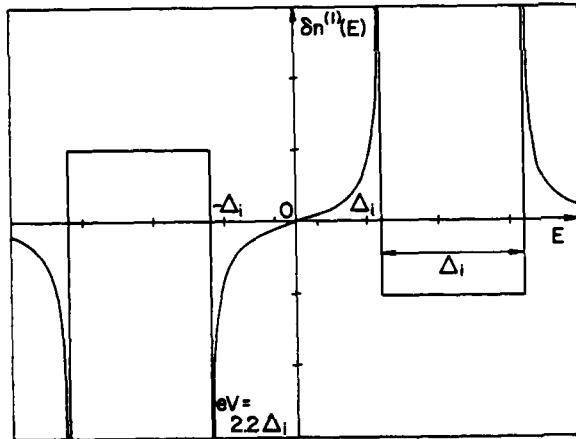


Fig. 1: The first order contribution  $\delta n^{(1)}(E)$  to the quasiparticle distribution function describing an excess number of excitations (particle-like for  $E > 0$  or hole-like for  $E < 0$ ) as a function of energy.

Consider the solid line in Fig. 1 for the case  $|eV| \sim \Delta_i + \Delta$ . We can qualitatively understand why two solutions of the gap equation may simultaneously be possible, as follows. Suppose that  $\Delta = \Delta_i$  is a self-consistent solution of the gap Eq. (1). Then the peak in  $\delta n(E)$  at  $1.1\Delta_i$  will be convoluted with the peak in  $N_1(E)$  at  $\Delta = \Delta_i$  and will give a large negative contribution to  $\chi$  in Eq. (2). It is thus plausible that there also exists a solution with  $\Delta > 1.1\Delta_i$  since in such a case the peak in  $\delta n(E)$  at  $1.1\Delta_i$  would not contribute to  $\chi$  and a more positive  $\chi$  is consistent with a larger gap (see Eq. (1)).

The effects of heating on  $\delta n(E)$  will in most instances introduce a large but smooth background. If spikes similar to those depicted in Fig. 1 show up on this background then clearly the above qualitative picture of why there may exist two gap solutions will remain valid. On the other hand gap enhancement depends very much on the level of the background.

The effect of nonequilibrium phonons becomes important when the phenomenological "escape" time  $\tau_{es}$  becomes much longer than the phonon scattering time  $\tau^{ph}$ , i.e., when  $\tau_{es} \gg \tau^{ph}$  where  $\tau^{ph}$  can be expressed as a function of  $\tau_0$  using the ratio of the normal electron and longitudinal phonon specific heats  $C_{el}/C_{ph}$  respectively<sup>9</sup>,

$$\tau^{ph}/\tau_0 = 35\zeta(3)C_{ph}/(4\pi^2 C_{el}).$$

Using the energy and number conserving approximation method of Eckern and Schön<sup>9</sup> we find that the effect of nonequilibrium phonons can be represented as simple heating when

$$1 \ll \tau_{es}/\tau^{ph} \ll (T/\Delta)(T/eV). \quad (5)$$

In such a case, the parameter  $\alpha$  becomes  $\alpha'$ .<sup>10</sup>

$$\alpha' = \alpha + \delta\alpha_h = \alpha - 2.59B\tau_0(eV/2T)^2(\tau_{es}/\tau^{ph}). \quad (6)$$

Note that  $\delta\alpha_h$  does not have any sharp step. By contrast,  $\chi$  computed with  $\delta n^{(1)}(E)$  has a step<sup>5</sup>. It is basically this step which leads to multiple solutions of the gap equation.

### 3. SIMPLE PREDICTIONS FOR EXPERIMENTS

#### 3.1 Qualitative Aspects

We quote a few qualitative features of the theory, some of which have already been experimentally observed, some of which will hopefully be checked in future experiments.

- a) The phase transition aspects of our theory have already been noted<sup>5,2</sup> and seem to agree with experimental findings<sup>6,7</sup>: at a certain voltage (see also Sec. 3.2), a low gap region (large injection current density) appears in the probe and grows relative to the larger gap region (low injection current density) as the total injection current increases. The expected hysteresis has also been observed<sup>6,7</sup>. This transition in the junctions is analogous to a liquid gas transition at constant pressure where the relative volumes are controlled by the total volume. The total injection current which controls the transition has no analogue in any of the other nonequilibrium first order phase transitions studied by ESSS.
- b) There is no threshold current, conductance or quasiparticle density in our theory. The two coexisting gaps can be in principle observed at the threshold voltage in many kinds of junctions, even at very low injection current density, as long as the energy difference between the two coexisting gaps (see following section) is large enough to be resolved from relaxation time smearing effects and other similar complications.
- c) The Chi and Clarke gap enhancement (to be published) and the coexisting gaps can be observed in principle in the same junction.

d) If heating effects are negligible, one of the two coexisting gaps may be enhanced with respect to the equilibrium gap.

e) The coexistence of a superconducting and a normal phase is possible, as can be seen using either the graphical methods of I and ESSS or an analytical method. Since this possibility is probably harder to see experimentally, we shall not give any more quantitative results on that matter.

Although the qualitative features described in paragraphs a) and b) have already been observed, observation of the phenomena described in paragraphs c), d), e) above would certainly give a stronger experimental support to our theory.

### 3.2 Quantitative predictions

Assume that Eq. (5) holds. Then Eq. (6) also holds. In the homogeneous case and when  $|eV|$  is very close to  $\Delta + \Delta_i$ , one can find a simple analytical form for Eq. (2). The transition voltage is determined using the procedure outlined in Eq. (4).

This leads us to our first quantitative prediction,

$$|eV_0| = \Delta_i + 1/2(\Delta_s(\alpha') + \Delta_l(\alpha')). \quad (7)$$

The transition voltage  $eV_0$  can be determined experimentally. It should be related to the injector gap and to the experimentally determined values of the two coexisting gaps  $\Delta_s(\alpha')$  and  $\Delta_l(\alpha')$  as described in Eq. (7). Note that Eq. (7) and Eq. (8) that follows are valid only when the condition

$$\frac{B\tau_0}{\alpha'} \left( \frac{\Delta_s(\alpha') + \Delta_i}{2T} \right) \pi \left( \frac{\Delta_i}{\Delta_s(\alpha')} \right)^{1/2} \ll 1$$

is satisfied.

The temperature and conductance dependence of the two coexisting gaps constitute our second quantitative prediction. If  $\Delta_i$  is roughly constant in the temperature range of interest and  $\Delta_i \gg \Delta_s(\alpha')$  we have

$$\Delta_l(\alpha') - \Delta_s(\alpha') \sim \frac{R^{-1}}{(T_c - T)^{3/4}} + \frac{R^{-2}}{(T_c - T)^{7/4}} \quad (8)$$

The last term depends on the square of the conductance ( $R^{-2}$ ) and on  $\tau_{es}$  and by assumption is smaller than the term proportional to the conductance.

### 4. CONCLUSION

Five qualitative predictions were given in Sec. 3.1 and two quantitative predictions in Sec. 3.2. Predictions concerning the absolute values of the observed gaps can be derived but they are less useful because they contain the parameter  $\tau_{es}/\tau_{ph}$  which may be harder to obtain experimentally. Nevertheless, it is useful to observe that our theory is not inconsistent with the fact that experimentally, the two coexisting gaps have values smaller than the equilibrium gap. If, as was the case in the theory presented in I,

the condition  $\frac{\tau_{es}}{\tau_{ph}} \left( \frac{\Delta_s(\alpha) + \Delta_i}{2T} \right) \ll \left( \frac{\Delta_i}{\Delta_s(\alpha)} \right)^{1/2}$

is realized, then the largest of the two coexisting gaps has a value larger than the equilibrium value. In practice, the above condition may not be satisfied.

It would be interesting to extend these ideas to low temperatures where a solution of the deterministic equation also leads to the possibility of a phase transition<sup>5</sup> but where a determination of the fluctuations, which we must know to determine the global stability, is more complicated than close to  $T_c$ .

If the predictions presented in this paper are verified, we are confident that our theory will also explain new experiments which have just been done<sup>11</sup>.

Finally, guided by the analogies between the generalized free energy determining the stability of the nonequilibrium state and the usual thermodynamic free energy, one can speculate that future experiments may exhibit phenomena analogous to spinodal decomposition in fluids<sup>12</sup>. A rapid change of voltage in a junction is analogous to a rapid change of pressure in the fluid. The current and junction conductance are analogous respectively to the volume and temperature of the fluid.

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